THE EFFECT OF OCCLUDED HYDROGEN ON THE RIGIDITY OF PALLADIUM AND THE POSSIBILITY OF THE EARTH'S CORE BEING SUCH AN OCCLUDING METAL

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By

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Submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy at New York University

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INTRODUCTORY STATEMENT

SEISMOLOGY HAS SHOWN quite conclusively that a definite core exists within the earth, its radius being roughly half that of the earth. The depth of the core has been shown by the reflection of seismic waves, much as the depth of the ocean is shown by the reflection of sound waves. The nature of the core presents a puzzle. The existence of tidal effects and the apparent absence of shear waves on the records of distant quakes were interpreted as indicating a core of little or no rigidity, and up to a decade or so ago a liquid core was quite generally held. Since then shear waves on the records of distant quakes have been tentatively identified, but their trace is very faint, indicating very little shear wave energy. The writer suggested (*Nature*, August 28, 1937) that the core might be a solid solution, i.e., a gas, presumably hydrogen heavily occluded by a metal. As a test of such an hypothesis it was proposed to measure the effect of such an occluded gas on the rigidity of the occluding metal. Palladium was selected because of its known readiness to absorb hydrogen. It was found that Koch, in Germany, had already done some work on the effects of occluded hydrogen on the elastic constants of palladium, but his results were very erratic. It was thought advisable to repeat Koch's work, using an entirely different and more refined method. A torsion pendulum was used as in Koch's case, but torsion was produced by an alternating magnetic field instead of mechanically, the period being obtained from the frequency at resonance. The specimen used was palladium wire .5mm. in diameter. Occlusion was measured by the resistance of the wire, using a Kelvin bridge. Occlusion and de-occlusion were produced by electrolysis. The torsion pendulum consists of a magnetized needle clamped to the end of the palladium wire. The alternating magnetic field is produced by a V. T. Oscillator. Length and radius of the wire are measured by micrometer microscope. The object is to obtain a curve, plotting rigidity against the amount of hydrogen occluded, and from this curve to see whether such a solid solution as core might satisfy the required values of rigidity for the earth's interior.

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The Effect of Occluded Hydrogen on the Rigidity of Palladium and the Possibility of the Earth's Core Being Such an Occluding Metal

When a disturbance is set up in an elastic solid, two waves are generated, a compressional wave and a shear wave. The former is due to change of volume, the latter to change of shape. From the theory of elasticity, we can express the velocities of these waves in terms of the elastic constants of the solid.

$$V_c^2 \text{ (Compressional)} = \frac{\lambda + 2\mu}{\rho}$$

$$V_s^2 \text{ (Shear)} = \frac{\mu}{\rho}$$

μ = Modulus of torsion

ρ = Density

λ = Lame's compression constant $= \dfrac{E\sigma}{(1+\sigma)\,(1-2\sigma)}$

E = Young's Modulus

σ = Poisson's ratio.

The earth being an elastic solid, these two waves are generated on a large scale in every earthquake, Professor Wiechert, of Gottingen, being the first to identify the two waves as such on earthquake records in 1899. Since we can express the elastic constants of the Medium in terms of these velocities,

$$\mu = \rho V_s^2$$
$$\lambda = \rho (V_c^2 - 2V_s^2)$$

it is clear that one of the best means of determining the elastic constants of the earth is by means of earthquake wave velocities.

Such determinations have been made by observers too numerous to mention, but in the process of such determinations, it became apparent that the earth, instead of being a body of continuously increasing density as we approach the center, was a body with frequent discontinuities. These discontinuities can best be represented by picturing the earth as made up of concentric shells, the density

and elastic properties changing sharply as we go from shell to shell. The shell that we are particularly interested in, in this discussion, is the shell existing about half way down in the earth and now known quite generally as the core of the earth.

The existence of the core was first suggested by Oldham in 1906 (1). Abundant confirmation of its existence has been given by the reflection of seismic waves from it and the refraction of seismic waves through it. The position of the core at 2920 km. below the earth's surface, as deduced by Gutenberg (2), is now generally accepted by seismologists. From the same source we find the radius of the core to be 3446 km. and the length of 1° on its surface to be 60.20 km. The effect of the core on compressional earthquake waves has been investigated rather thoroughly. Due to refraction of the waves by the core a well known shadow zone exists on the earth's surface from about 105° to 142° from the epicenter of the quake. Within this belt about 37° wide, seismographs are practically screened from compression earthquake waves. At the end of the belt a strong focussing effect is observed. After being eclipsed throughout this 37° zone, the waves suddenly reappear on seismographs near 142° from the quake with an energy even greater than that at the disappearing point, indicating an apparent focussing at 142°. To account for this, Gutenberg (2) computed the velocity of earthquake compressional waves as increasing with depth from 8 km. per sec. at the surface to 13.7 km. per sec. at the core. Here at the discontinuity he postulated a sudden drop from 13.7 km. per sec. to about 6 km. per sec. In the core again he postulated a gradual rise in velocity from 8 km. per sec. to 11.5 km. per sec. at the center of the earth. While the shadow zone and the focussing effect just mentioned are quite pronounced, compressional waves have, however, been observed as advancing into this shadow zone beyond 105°. These waves have usually been described as waves diffracted around the core. These diffracted waves are of long period (over 10 secs.) and small amplitude, the latter decreasing with increasing distance. More recently compressional waves have been observed as creeping back into the shadow zone at the other end. They have been observed back from 142° as far as 110°. Wiechert, in an unpublished paper, in 1922, attributed these waves likewise to diffraction, but no attempt was made to explain the mechanism of such diffraction. The waves diffracted back into the shadow zone at its end, differ both in period and amplitude from those diffracted forward into it from its begin-

ning, the period being shorter and the amplitude larger. In view of this difference which seems to indicate a difference of origin and in the absence of any suggested possibility of the mechanism of such a diffraction process, Miss Lehmann (3) suggested that these back-diffracted waves were due, not to diffraction but to refraction due to a discontinuity within the core. She calculated that a velocity of 10 km. per sec. in the mantle outside the core dropping suddenly to 8 km. per sec. in the outer part of the core and rising to 8.6 km. per sec. in the inner core, would explain the observed facts, the radius of this core being 1400 km. Gutenberg (2) accepts Miss Lehmann's suggestion of refraction instead of diffraction for these formerly named diffracted core waves but disagrees with her on the velocity distribution required. He postulates a velocity of about 8 km. per sec. at the inside surface of the core increasing to about 10.2 km. per sec. 1950 km. deeper, and then a rapid increase with depth, the velocity increasing to 11.4 km. per sec. within 300 km., then decreasing slowly to 11.3 km. per sec. at the center.

These and similar investigations while showing conclusively that a core exists, and while throwing light on such properties as its density, tell nothing at all about the state of the core, i.e., whether it is solid or fluid. The only seismic tool which can throw any light on the state of the core is the shear wave. Prior to the advent of seismology, a fluid core was generally held.

In the light of the widely accepted fluid origin of the earth, it was quite generally held that solidification had probably not yet proceeded all the way to the core and that the core was therefore liquid. The action of the tides supports this view. The magnitude of the lunar tides calls for an average rigidity in the earth less than that found in the crust. Jeffreys (4) postulates zero rigidity in the core to balance the high rigidity of the crust. The period of precession of the earth also supports the view of a liquid core. Calculated for a perfectly rigid earth, the Eulerian period of precession would be 300 days (5). The only such observed period is the period of 427 days observed by Chandler, as measured by variation of latitude (6). Hough (7), discussing the Chandler period, computes that the Eulerian period for the earth would be lengthened to 440 days on the assumption that the earth had an average rigidity equal to that of steel. With the advent of seismology, further evidence for a liquid core was forthcoming. While the compression wave after disappearing into the shadow zone at 105°, reappeared

with renewed vigor at 142°, the shear wave after similarly disappearing at 105°, failed to reappear—at least, observers failed to find it. This seemed to indicate that the core was incapable of transmitting a shear wave. Hence, many concluded that the core had zero rigidity, and hence was probably a liquid. It is possible for a shear wave to be generated in a liquid, but it is damped out within a distance of a wave-length. Seismological evidence seemed, therefore, to confirm the existing theory of a liquid core. However, an alternative suggestion was offered in Japan to account for the apparent failure of shear waves to penetrate the core. Investigating the effect of the sharpness of a discontinuity on the reflection and refraction of earthquake waves, Sezawa and Kanai (8) showed that where a zone of finite thickness separates two media of different density and rigidity, the wave-length is a deciding factor in transmission or reflection. If the incident wave-length is long in comparison with the thickness of the intervening zone, the amplitude of the reflected wave will be large, while that of the transmitted wave is small. If, however, the incident wave-length is small in comparison with the thickness of the separating zone, the amplitude of the transmitted wave will be large, while that of the reflected wave will be small. Since shear waves are, in general, of longer wave-length than compression waves, Sezawa and Kanai suggested that the failure of shear waves to emerge from the core may be attributed to this reflection which the waves would meet with, both when they enter the core and for those which find their way in also, when they leave it, so that practically none would escape. Further weight would seem to be given to this suggestion of Sezawa and Kanai by the discovery of Dahm (9) that a transition layer exists between core and mantle, though the thickness of this layer suggested by Dahm, 220 km., would be a little too great for the findings of Sezawa and Kanai.

So far it has been taken for granted that the shear wave does not emerge from the core, in which case we have a choice of explaining its non-emergence either by lack of rigidity in the core or by total reflection.

As seismic investigation has progressed, however, evidence has been accumulating that the shear wave does penetrate and emerge from the core albeit with small energy. Macelwane (10), (11) found what he thought were core shear waves in the South Pacific Earthquake of 1924. Imamura (12) made a similar identification,

as also did Krumbach (13), while Bastings (14), after a thorough study of the New Zealand Earthquake of 1929, seems quite positive in his identification of such waves, writing in his conclusion, "The rigidity of the core, at least for vibrations of short period, thus seems to be definitely established." Here, then, we have four independent investigators in widely different parts of the globe making exhaustive studies of different quakes and claiming to have identified at least feeble core shear waves. It would seem, then, that the present evidence from seismology would indicate at least some rigidity for the core, and hence that it is not liquid.

Macelwane and Gutenberg writing different chapters of the latest book (15) dealing with the interior of the earth, after weighing the evidence, refuse to make definite pronouncement on the nature of the core and leave it, for the present, an open question. Gutenberg writes, "The problem of whether the core of the earth is solid or fluid is still open. Though most specialists in this field agree that all observations indicate a large drop in rigidity at the boundary of the core, no final conclusions have yet been reached as to whether the rigidity and the viscosity in the core are so small that we would call the material fluid under normal conditions or solid." Macelwane, after stressing the difficulty of observing core shear waves due to the fact that they are superposed on the faster and still incoming compression waves, continues, "In view of the disagreement of other competent seismologists, the inherent difficulties of observation and interpretation, and the incomplete and unsatisfactory state of mathematical wave theory, and of our knowledge of waves in general, the final and definite decision as to the reality of shear waves in the core must be left to the future. But in the meantime, seismological evidence must not be discounted or rejected because of presumptively contrary evidence from other fields, which, on careful analysis, is found to be subject to even greater uncertainties."

Gathering together all the evidence, it seems highly probable that the core has some rigidity, but the small energy of the emerging shear waves seems to indicate that it is slight. If it has any rigidity it cannot be a liquid. One might appeal for the low value of the rigidity to the high temperature that is supposed to exist at the center of the earth, the general effect of increasing temperature being to lower the rigidity (16). But against this, one would have to say first that any such lessening of rigidity by temperature would be

more than offset by the increase due to pressure; and secondly, as Gutenberg (2) has pointed out, the temperature at the center of the earth is probably not as high as has generally been supposed. A different reason altogether was advanced by the writer in 1937, to account for the low rigidity of the core (18). Neither the reflection explanation of Sezawa and Kanai nor a liquid core seemed to explain all the facts. Sezawa's explanation made no mention of lower rigidity and therefore offered no explanation of the low rigidity required for tidal action. A liquid core, on the other hand, was hard to picture in view of the discontinuity required in the core by Gutenberg and Lehmann and the apparent identification of core shear waves by four different observers. Hence, the writer suggested that the core was neither solid nor liquid but a solid solution, i.e., a gas, probably hydrogen, heavily occluded by a metal. It is well known that some metals, after absorbing some thousand times their volume of hydrogen, show no apparent effects of the absorption, and seem in all visible respects like the ordinary metal. One would naturally expect, however, that an elastic constant such as rigidity must be in some way effected by the occluded gas. The thought was that the occluded gas would lower the rigidity to allow shear waves to pass through, but with little energy. As a first test, therefore, of the hypothesis of a solid solution core, it was proposed to measure the effect, if any, of an occluded gas on the rigidity of the occluding metal. Palladium was selected as the metal because of its known readiness to absorb hydrogen. On looking through the literature on occlusion, it was found that Koch (19) had already done some work on the effects of occluded hydrogen on the elastic constants of palladium, but his results are very erratic, showing, however, a definite decrease in rigidity as hydrogen was absorbed. It was thought advisable to repeat Koch's work, using an entirely different and more refined method. Koch had used a long torsion pendulum with a meter length of palladium wire as the suspension. The wire was enclosed in a cylindrical glass tube almost the length of the wire and several centimeters in diameter. The tube was filled with sulphuric acid, occlusion being produced electrolytically. The wire itself formed one electrode, the other consisting of metal strips on the inside of the glass cylinder. Torsion was produced mechanically by giving an impulse to the lower end of the wire. The period was measured by stop-watch, and measurements of length and radius of the wire were made through the glass from the outside. The

amount of hydrogen occluded was measured by the resistance of the wire, Fischer (20) having previously worked out the relation between the amount of hydrogen occluded and the electrical resistance. The writer used a torsion pendulum, as did Koch, but torsion was produced by an alternating magnetic field instead of mechanically, the period of the pendulum being obtained from the frequency at resonance. The pendulum consisted of a 3–5 cm. length of palladium wire half a millimeter in diameter, to the end of which a magnetized needle is clamped. The alternating magnetic field, producing torsion of the pendulum is generated by a vacuum tube oscillator, the current from which, after passing through an amplifier, is fed into a large choke coil with a 1½-inch removable soft iron core. The pendulum is set up in front of the core so that the needle is on a line with its diameter. At resonance the end of the needle vibrates with an amplitude of about half an inch. A General Electric clock is placed in parallel with the core and the frequency measured over a period of ten minutes by comparison with a stopwatch, each second of the clock being equal to 60 cycles of the pendulum. Consecutive readings of ten-minute intervals showed agreement within one-tenth of a second. In determining rigidity, the formula

$$\mu = \frac{8\pi I l}{r^4 T^2}$$

was used, where

μ = rigidity
I = Moment of Inertia of needle and chuck
l = length of pendulum
r = radius of wire
T = period of oscillation

I was determined by attaching to the chuck holding the needle, a small symmetrical copper cylinder of known moment of inertia, using the formula

$$I = \frac{I' l'}{T'^2} \cdot \frac{T^2}{l}$$

The length of the pendulum l was measured by a traveling microscope reading to .002 cms. A Gaertner micrometer eyepiece reading to .005 mm., was used to measure the diameter of the wire. A

Kelvin bridge was used to measure resistance. Fischer (20) having already worked out the formula connecting resistance with volume of gas occluded,

$$\frac{W}{W_o} = a + b\,H$$

w = resistance
H = volume of gas occluded
a = 1.0292
b = 0.000,668

Fischer points out that the formula is reliable only between the limits of 30–950 volumes of gas occluded, i.e., between the points where the wire has 30 times its volume of gas and where it has 950 times its volume. Between these two points the curve obtained by plotting resistance against volume of hydrogen occluded is a straight line, beyond them at either end a slight curvature appears.

As a preliminary experiment it was proposed to find out by how much the rigidity might be expected to vary. A piece of new palladium wire was taken and a current of 10 amperes passed through for a few seconds to drive out any hydrogen that might be in the wire. The rigidity increased 1.7%. Current of the same value was again passed through, this time for one minute. The rigidity was found to increase a further 8.4%. The wire was then occluded in a graduated electrolysis apparatus, where the volume of occluded hydrogen was measured by the difference between the columns of oxygen and hydrogen that formed. Four hundred times its volume of hydrogen were occluded by the wire, and the rigidity was found to decrease 16.6%. Current was then again passed through to drive out this hydrogen and the rigidity increased 22.8%. The wire was again charged with 450 times its volume of hydrogen, and the rigidity decreased 21%. The average decrease in rigidity for a wire that was saturated, i.e., a wire that was given hydrogen until it ceased to absorb any more, was found to be about 13.7%. The pendulum was placed under a bell jar and the period measured in vacuo, in hydrogen, and under pressure, but no measurable differences of period were observed. The occluded hydrogen was found to leak out always when the wire was left standing, but the leakage was slow and quite negligible during the duration of an experiment.

Results

The results have been put down in the order in which they were determined, not in the order of ascending or descending rigidity or resistance. The procedure was to take a piece of unoccluding wire, measure its resistance and determine its rigidity. Then the wire was made to occlude hydrogen electrolytically, the amounts of occlusion varying widely. Resistance and rigidity would again be determined. Then further occlusion or de-occlusion, according as points were needed to fill gaps on the curve.

For non-occluding palladium, Koch found values of rigidity varying from 4.112 to 5.177 x 10^{11} dynes cm^2. The author's value for the rigidity of non-occluding palladium is 5.156+.015 x 10^{11} dynes cm^2. In plotting the curve showing change of rigidity with occlusion, it was thought best to plot rigidity against resistance rather than against volume of occluded gas. The reason was two-fold. First, the quantity actually measured was the resistance, and hence the curve really obtained was a rigidity resistance curve. As Fischer himself pointed out, his formula for converting resistance into volume of occluded gas does not hold for less than 30 volumes or more than 950 volumes. Hence the initial and final portions of the resistance curve, after translation into a volume curve would be invalid. Moreover, the writer on constructing his own curve for converting resistance into volume of gas occluded, obtained as Fischer did, a straight line curve for the region measured, but the slopes of the curves do not agree. It was thought best, therefore, to avoid any ambiguity arising from conversion of resistance into occluded volume, and to plot rigidity against resistance. On the basis of the author's conversion curve, the rigidity resistance curve presented here would represent the change of rigidity from zero occlusion up to about 700 volumes, i.e., an amount of occluded gas equal to 700 times the volume of the unoccluded wire. The ready escape of hydrogen, when heavily occluded, made rigidity determinations more uncertain as occlusion progressed. For this reason no attempt was made to go beyond 700 volumes and the latter part even of this curve is perhaps unreliable.

It was not the purpose of this paper to discuss the mechanism by which the absorbed hydrogen lowers the rigidity of the absorbing metal. It might be said in passing, however, that since it is generally accepted (21), (22), that the hydrogen occlusion in the case of palladium is a lattice occlusion producing lattice expansion, the

lowering of the rigidity would seem to be due to the lessening of the electrostatic repulsion of the ions consequent on such expansion. We are here assuming Brock's contention (23), that the rigidity at least of the alkali metals is due solely to electrostatic repulsion of the ions.

Resistance x 10^{-5} ohms	Rigidity x 10^{11} dynes/cm^2
1304	5.127
1841	4.770
1332	5.050
1327	5.113
2100	4.595
1880	4.761
1605	4.748
1345	5.132
1326	5.270
1325	5.182
1585	5.021
1584	4.927
1767	4.875
1760	4.973
1740	4.988
1898	4.930
2074	4.847
2068	4.636
2308	4.402
2343	4.457
1337	5.199
1344	5.196
1657	4.933
1646	4.963
1722	4.829
1567	5.025
1665	4.986
1814	4.815
1916	4.775
2301	4.661
2312	4.520
2252	4.623
2213	4.580
1344	5.163

Resistance x 10^{-5} ohms	Rigidity x 10^{11} dynes/cm^2
2304	4.408
2210	4.432
2252	4.552
1814	4.850
1665	4.953
1344	5.155
1345	5.158
1749	4.859
2042	4.562
1347	5.078
2285	4.535
1337	5.096
1344	5.196
1585	5.039
1585	4.927
1665	4.953
1916	4.736
2218	4.389
2050	4.455

By way of anticipating one or two questions of detail, all of the determinations were made with specimens cut from the same wire. The wire in question had a guaranteed purity of 99.5% or better. No attempt was made to experiment with other samples since this was considered more than pure enough to show the desired relationship between rigidity and occlusion. No attempt likewise was made to determine what, if any, residual gases might be in the wire at the start. As mentioned in the text, a slight amount of hydrogen is probably present in all palladium, but this slight initial amount would not affect the general slope of the curve showing increasing occlusion against rigidity. An interesting visual proof that the gas occluded during the experiments was hydrogen, was afforded by making a piece of the occluding wire the anode in an enclosed electrolysis outfit. While hydrogen bubbled off profusely from the cathode, no gas at all appeared at the anode (our occluding wire). The oxygen released by electrolysis combined at once with the occluded hydrogen to form water and no oxygen appeared until this occluded hydrogen had been used up.

With regard to the accuracy of the results obtained, it should be pointed out that the most difficult quantity to measure was the

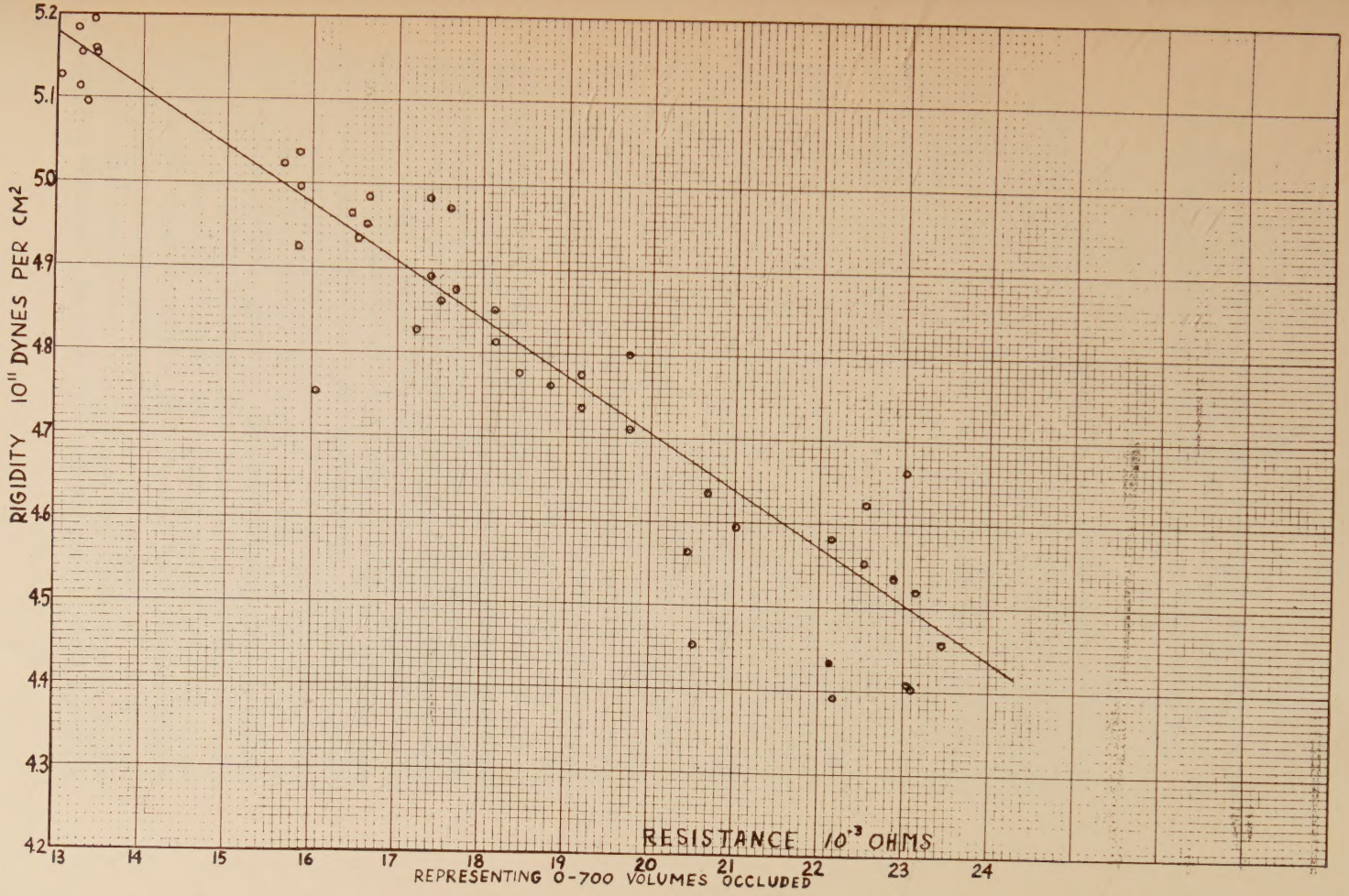
diameter of the wire. A micrometer screw was tried but was ruled out because of its pinching effect on the wire. A micrometer microscope reading to .005 mm. was finally used. Higher magnification was impractical because of the depth of field of a wire half a mm. thick. In this depth of field lay the chief source of error. It was extremely difficult to judge just when the two edges of the diameter of the wire were in focus and since the radius appears to the fourth power in the formula $\frac{8\pi I l}{r^4 T^2}$, this error completely masks all others. At the same time, since the chief measurable agent by which the occluded gas changed the rigidity, was the diameter of the wire, it was necessary to have a magnification at least as high as this to measure the extremely slight changes of diameter involved. The present arrangement is not to be recommended, therefore, for determining the absolute value of the rigidity of palladium. For this it would be better to use a wire of larger diameter and micrometer screw. The figures obtained by the author for the rigidity of unoccluding palladium can be relied on to within 0.3%, but this precision is too high for determinations made on an occluding wire, since another factor now appears. The slightly irregular way in which the wire absorbs hydrogen introduces a still further error in the measuement of the diameter. Continual escape of hydrogen made a repetition of any determination impossible, hence it was impractical to express the precision of each determination. The method used would warrant a precision of 0.3%, but the irregularities of occlusion increase this error, so that the points are scattered fairly widely about the adopted curve.

The discrepancy between the author's resistance-occlusion curve and that of Fischer may possibly be due to different specimens of palladium used. The author's was 99.5% pure, but Fischer states nothing about the purity of his specimen. Slight discrepancies also occurred between Fischer's curve and those of contemporary and antecedent workers, but no reason has been advanced by way of explanation. The slight discrepancy, however, whatever its explanation, in no way invalidates the conclusion of the thesis.

Conclusion

Hydrogen when occluded by palladium greatly lowers the rigidity of the metal. The loss amount to about 15% for an occlusion of 700 volumes, but as much as 30% loss has been observed for higher

occluded volumes. The curve that was obtained relating change of rigidity with occlusion is a straight line and seems to indicate that with continued occlusion, the rigidity would continue to decrease. The escape of the occluded hydrogen makes rigidity determinations difficult when large volumes are re-occluded. In the core of the earth, the enormous pressure would prevent such escape, hence it is plausible to imagine a metallic core occluding sufficient hydrogen to reduce the core's rigidity practically to zero. On the basis of the present work, therefore, a solid solution is proposed as representing the state of the earth's core. The gas presumably would be hydrogen, the metal one of the many metals known to occlude hydrogen readily. Such a solid solution core would have the advantage over a liquid core that it would permit feeble shear waves to pass through, as seismic investigation seems to require. It would have the advantage over a solid core that it would have the low rigidity demanded by tidal action and the Eulerian period.


RIGIDITY 10^{11} DYNES PER CM²
5.2
5.1
5.0
4.9
4.8
4.7
4.6
4.5
4.4
4.3
4.2
13
14
15
16
17
18
19
20
21
22
23
24
RESISTANCE 10^{-3} OHMS
REPRESENTING 0-700 VOLUMES OCCLUDED